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OFFICE OF NAVAL RESEARCH
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Technical Report No. 11

PHYSICAL CHEMISTRY OF THE DEEP OCEAN ENVIRONMENT

by
R. A. Horne

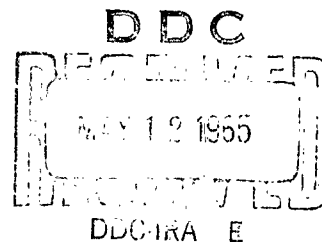
Prepared for presentation before the
Marine Sciences and Research Section of the
Second U.S. Navy Symposium on Military Oceanography

Washington D.C., May 1965

Arthur D. Little, Inc.
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May 1, 1965

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PHYSICAL CHEMISTRY OF THE DEEP OCEAN ENVIRONMENT

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R. A. Horne

Arthur D. Little, Inc., Cambridge, Massachusetts 02140

INTRODUCTION

Naval operations occur above, on, and in a medium which is easily one of the most complex and ill-understood substances known to man - sea water. The concurrence of relatively low temperatures (-1 to $+4^{\circ}\text{C}$) and high hydrostatic pressures (up to about $15,000 \text{ lbs/in}^2$) in the ocean depths further complicates the situation and makes the deep ocean perhaps the most extraordinary environment in our universe. This paper reviews the results of some simple experiments which we have performed in our laboratory over recent years in a simulated deep ocean environment (Fig 1).

THE STRUCTURE OF SEA WATER*

There are several theories of the structure of water. There is also a minority opinion which insists that water has no structure at all. But at the present time in the United States the most popular model for the state of affairs in pure water appears to be that of Frank and Wen (1957), who proposed that water consists of transient "clusters" of more-or-less ordered, H-bonded water (sometimes, perhaps unfortunately, referred to as "ice-like") swimming in and in equilibrium with "free", monomeric water (Fig 2). Nemethy and Scheraga (1962) have calculated the size and concentration of these clusters as a function of temperature. The application of pressure or the addition of an electrolyte also affect the extent of water structure.

Roughly speaking sea water is an 0.5M NaCl solution with a trace of MgSO_4 . The addition of electrolytes further complicates the structure of the system. If an ion, such as Na^+ , is placed in water the coulombic attraction between the ion's charge and the partial charges of opposite sign of the water dipoles results in a crowding or "electrostriction" of the water molecules about the ion and an overall volume decrease (Fig 3). If the spacial distribution of water molecules in the first solvation sheath of an ion is symmetrical, such as $\text{Na}(\text{H}_2\text{O})_4^+$, the ion will be a structure-maker and will tend to make the surrounding water more ordered as reflected in an increase in

* For a more detailed discussion of the physical chemistry and structure of sea water see R. A. Horne, J. Water Resources Res., in press, 1965.

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relative viscosity (Fig 4) whereas if the first hydration sheath is asymmetrical, such as $K(H_2O)^+$ or $Cl(H_2O)^-$, the ion will be a structure-breaker and will tend to make the surrounding water less ordered as reflected in a decrease in relative viscosity (Fig 4) (Horne, 1964). $Mg(H_2O)_6^{++}$ $SO_4(H_2O)_{12}^-$, it is interesting to note, like many other 2:2 electrolytes is a very powerful structure-maker (Fig 4).

THE ELECTRICAL CONDUCTIVITY OF SEA WATER

Electrical conductivity, Λ , can be treated as a rate process and its activation energy, E_a , calculated from the integrated form of the Arrhenius equation

$$(1) \quad E_a = (\log \Lambda_2 - \log \Lambda_1) 4.576 T_2 T_1 / \Delta T$$

where the T's are absolute temperatures. $E_{a,cond.}$ for sea water exhibits a maximum (Fig 5) and for chlorinities less than about 10 ‰ the maximum occurs at the temperature of maximum density (Horne, 1964). The rate-determining step of the "normal" electrical conductivity of aqueous solutions of strong 1:1 electrolytes is "hole" or vacancy formation in the solvent. The process occurs both in the free and clustered water, and the decrease in $E_{a,cond.}$ with decreasing temperatures at temperatures below the maximum reflects the fact that "hole" formation is facilitated by the increasing amount of less dense, more open, clustered water (Horne, 1964 and Horne and Courant, 1964).

We have measured the effect of pressure on the electrical conductivity of sea water (Horne and Frysinger, 1963). In the range of oceanographic interest, pressure, like temperature, decreases $E_{a,cond.}$ indicating that its application tends to disorder liquid water (Horne, Myers and Frysinger, 1963). We found that Walden's rule

$$(2) \quad \Lambda^\circ \eta^\circ = \text{Constant}$$

where η is the viscosity is a good approximation for sea water under the condition of varying temperature but not varying pressure (Horne and Courant, 1964). However, we found that we were able to curve fit the observed pressure-dependence of the electrical conductivity of sea water by simple corrections for (1) the increased dissociation of $MgSO_4$ (Fisher, 1962), (2) the compressibility, and (3) the square of the viscosity (Horne and Courant, 1964). The apparent dependence on η^2 is an artifact arising from the reduction of the radii of hydrated ions by pressure (Horne, 1963) - a phenomenon first noted by Zisman (1932). The structural effects of pressure differ in one very important respect from those of temperature: temperature breaks up the structure of the bulk water but does not greatly affect the local water structure near ions, pressure, on the other hand, destroys both the bulk structure and the hydration atmospheres of ions.

THE VISCOSITY OF SEA WATER

On the basis of what now appears to have been a superficial similarity of their activation energies electrical conductivity, viscous flow, self-diffusion, and dielectric relaxation in water were believed to all involve the same rate-determining step (Glasstone, Laidler and Eyring, 1941, and Wang, Robinson and Edelman, 1953). However, more detailed examination, especially in the 0 to +10°C region reveals important differences. Miller (1963) reports no discontinuities in $E_{a,vis.}$ at 4°C in the viscosity of water and our recent measurements (Horne, Courant, Johnson, and Margosian, 1964) on both pure water and sea water yield the same result. $E_{a,vis.}$, unlike $E_{a,cond.}$ shows no maximum (Fig 5) hence the mechanism of viscous flow is different from that of normal conduction, possibly involving some kind of rotational tumbling of the clusters through the free water.

At the present time we are just completing an experimental study of the effect of pressure on the viscosity of pure water and sea water. The data have not been analyzed yet in detail, but qualitatively they show the same phenomena revealed by the much earlier work of Cohen (1892) on aqueous NaCl solutions, namely an initial decrease in viscosity with increasing pressure due to the destruction of the structure in liquid water and then, after the order has been destroyed, an increase in viscosity with increasing pressure as observed for "normal" non-associated liquids.

THE ELECTRICAL CONDUCTIVITY OF AQUEOUS MAGNESIUM SULFATE SOLUTIONS

Earlier we mentioned that $MgSO_4$ is a very powerful structure maker. $E_{a,cond.}$ of aqueous $MgSO_4$ solutions (Horne and Johnson, 1965) has a temperature dependence similar to that of viscous flow rather than normal ionic conduction (Fig 5). Evidently $MgSO_4$ is so highly hydrated that its species are too large to fit into the "holes" and as a consequence the mechanism of their transport in aqueous solution resembles that of the water clusters.

MISCELLANEOUS STUDIES

In addition to fundamental research on the physical-chemistry of sea water we have also turned our attention to problems of more immediate practical consequence. Our interest in marine corrosion at great depths prompted us to examine the effect of pressure on the dissociation constant of the chloride complex of a constructional metal.

(3)



Due to electrostriction (see above) the application of pressure tends to force this reaction to the right increasing the dissociation constant by about 20 fold in going from 1 to 2,000 atmospheres (Horne, Myers and Fry-singer, 1964). The performance of unshielded electro-chemical devices such as batteries on the ocean floor has been of concern to us (Horne, 1963) and

we have examined the effect of hydrostatic pressure on the electrical conductivity of some typical battery electrolytes (see Table 1) (Horne, Bannon, Sullivan and Frysinger, 1963). Notice the relatively large effect in the case of sulfuric acid due, in part, to the increased dissociation of bisulfate ion by electrostriction (Horne, Courant and Frysinger, 1964).

(4)

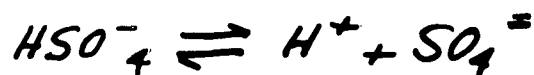


Table 1

Effect of Pressure on the Conductivity of Battery Electrolytes

<u>Battery Electrolyte</u>	<u>% Increase in Elect. Cond. at 1,000 atm. at 25°C</u>
Potassium Hydroxide	1.5
Sulfuric Acid	5
NaCl enriched with MgCl_2	2

Measurements of voltage versus current for button-type Ni-Cd and Leclanche cells under simulated deep ocean conditions yielded results that still leave us baffled. We expected the structure-breaking properties of pressure to decrease the structure at the electrolyte-electrode interface thereby facilitating the transport of ions and decreasing polarization, however, contrary to expectation, polarization appears to worsen with increasing pressure (Fig.6). We hope to look into this matter further (for studies of the performance of lead-acid batteries under pressure, see Schumaker, 1961).

Certain marine sedimentary materials such as montmorillonite have exchangeable cations. The effect of pressure on ion exchange equilibria had never been previously examined. Needless to say, we could not resist the temptation of applying our equipment to this purpose. We found little effect of pressure on the K^+/H^+ exchange on a synthetic, strong, sulfonic acid type, cation exchange resin, however, in the case of the $\text{Sr}^{++}/\text{H}^+$ exchange as pressure increases the resin's preference for Sr^{++} increases (Horne, Courant, Myers and George, 1964). Evidently the pressure-produced dehydration of the more heaving hydrated Sr^{++} ion is more marked than for the proton, hence the charge density (and thus the coulombic attraction for the resin sites) increases more steeply with increasing pressure for Sr^{++} than for H^+ .

CONCLUSION

Today one frequently hears of the application of the specialized knowledge of the various disciplines to marine science. This certainly need not be a one-way exchange of advantage. I hope that the experiments I have discussed

will provide an illustration of how what began as an interest in the simulation of the deep ocean environment in due course lead to fundamental research in solution physical chemistry. Oceanography has reopened, not a new, but a neglected field of physical chemistry, a field which I am convinced will prove to be a most fascinating and fruitful one - the effect of pressure on aqueous electrolytic solutions.

ACKNOWLEDGEMENT

This work was supported in part by the Office of Naval Research.

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Figure 3 Decrease in Water Volume Produced by Electrostriction

Figure 4 The Effect of Electrolyte Addition on the Relative Viscosity of Water

Figure 5 The Temperature-Dependence of the Activation Energy of Various Transport Processes in Water

Figure 6 Discharge Characteristics of an Eveready Size AA Dry Cell at 25°C Under Pressure

Table 1 Effect of Pressure on the Conductivity of Battery Electrolytes

Figure 1

High Pressure Equipment

(Manufactured by Harwood Engineering Company, Inc.)

for Simulating the Deep Ocean Environment

(-10 to +50°C, 15 to 200,000 lbs/in²)

A

Front View showing (on the left) the precision Shedlovsky-type conductivity bridge, (below) the thermostatic bath control, (above, center) the Foxboro "Dynalog" recorder which is connected to the maganin pressure cell, and (right) the cabinet housing the high pressure producing equipment with the hand pumps in the foreground.

B

Rear View showing (extreme left) the pressure intensifier, (above it and to the right) the main check valve, (left foreground) the cooling unit, (center) the thermostatic bath with the top of the pressure vessel showing, and (right) the potentiometer used in conjunction with measurements of the bath temperature.



Figure 2

Schematic Diagram of the Frank-Wen Water Model

(Nemethy and Scheraga, Journal of Chemical Physics,
v. 36, p. 3382. 1962.)

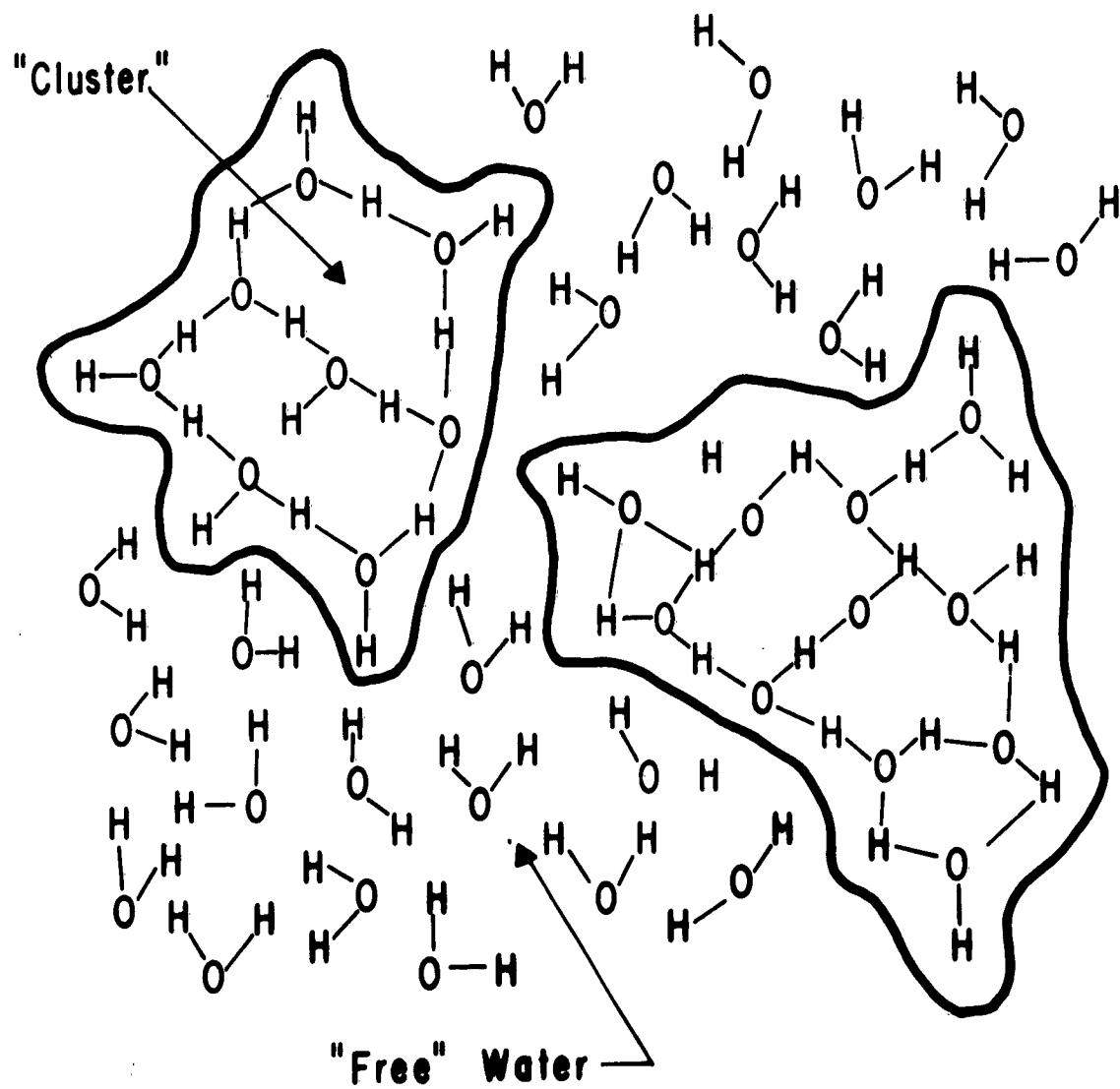


Figure 3

Decrease in Water Volume Produced by Electrostriction

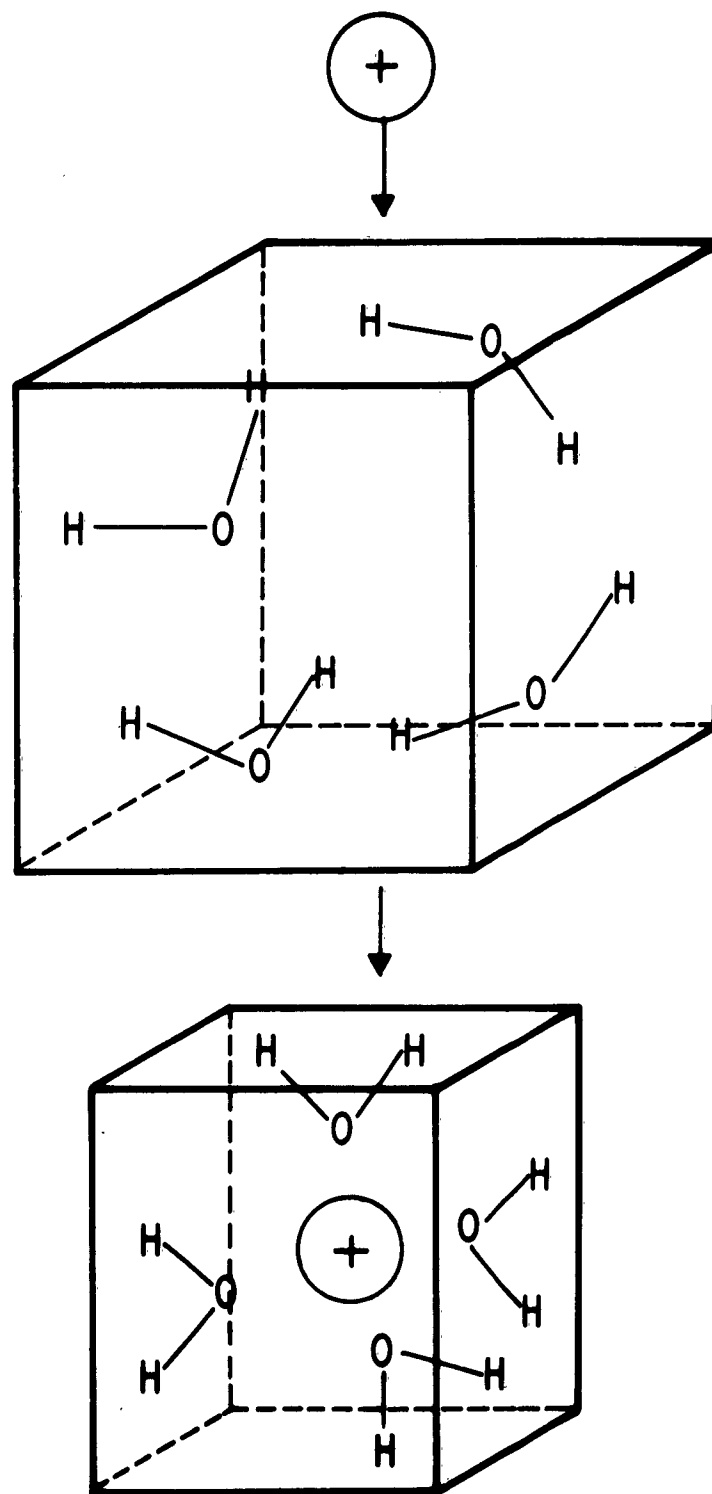


Figure 4

The Effect of Electrolyte Addition on the Relative Viscosity of Water

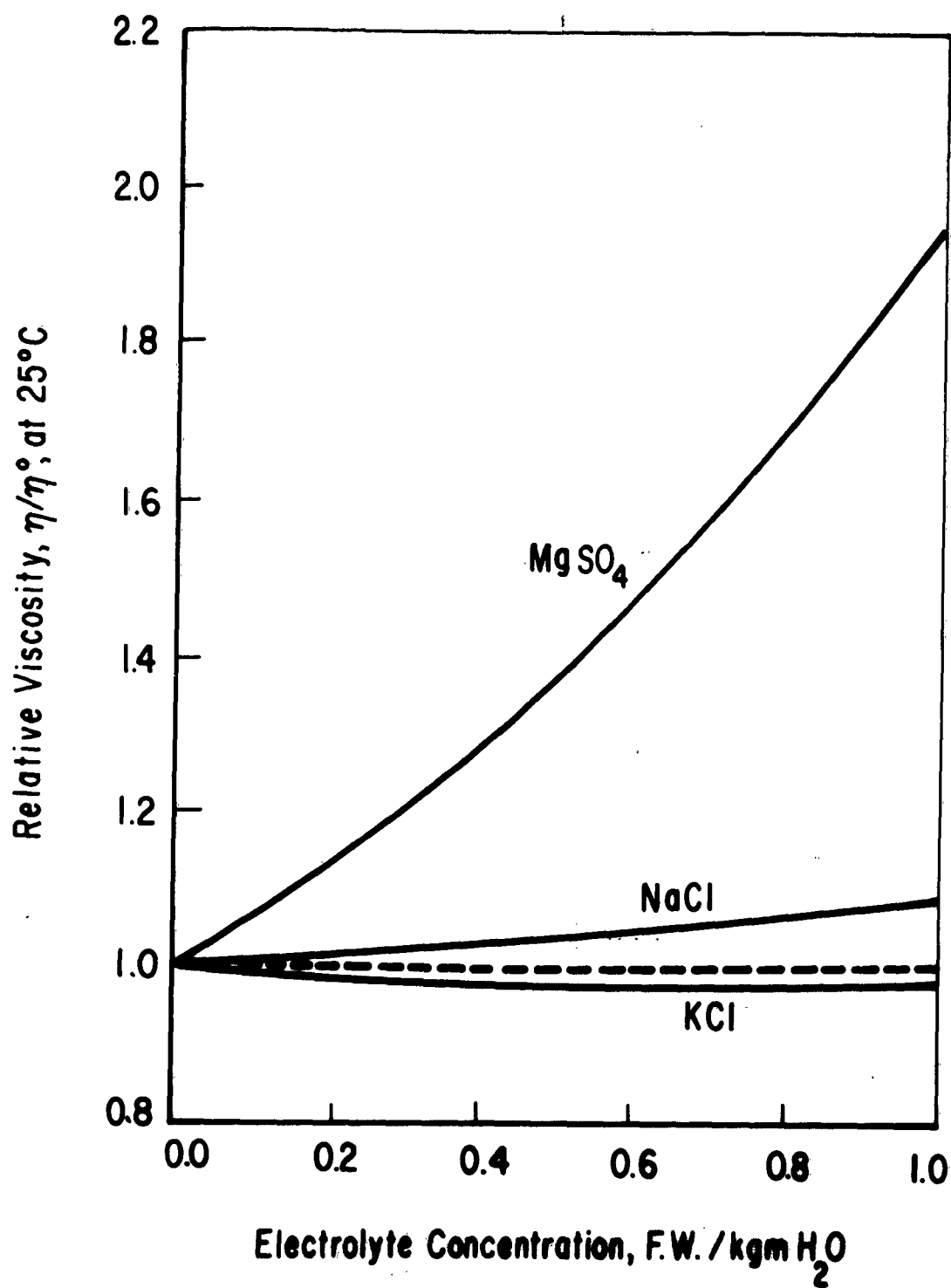


Figure 5

The Temperature-Dependence of the Activation Energy
of Various Transport Processes in Water

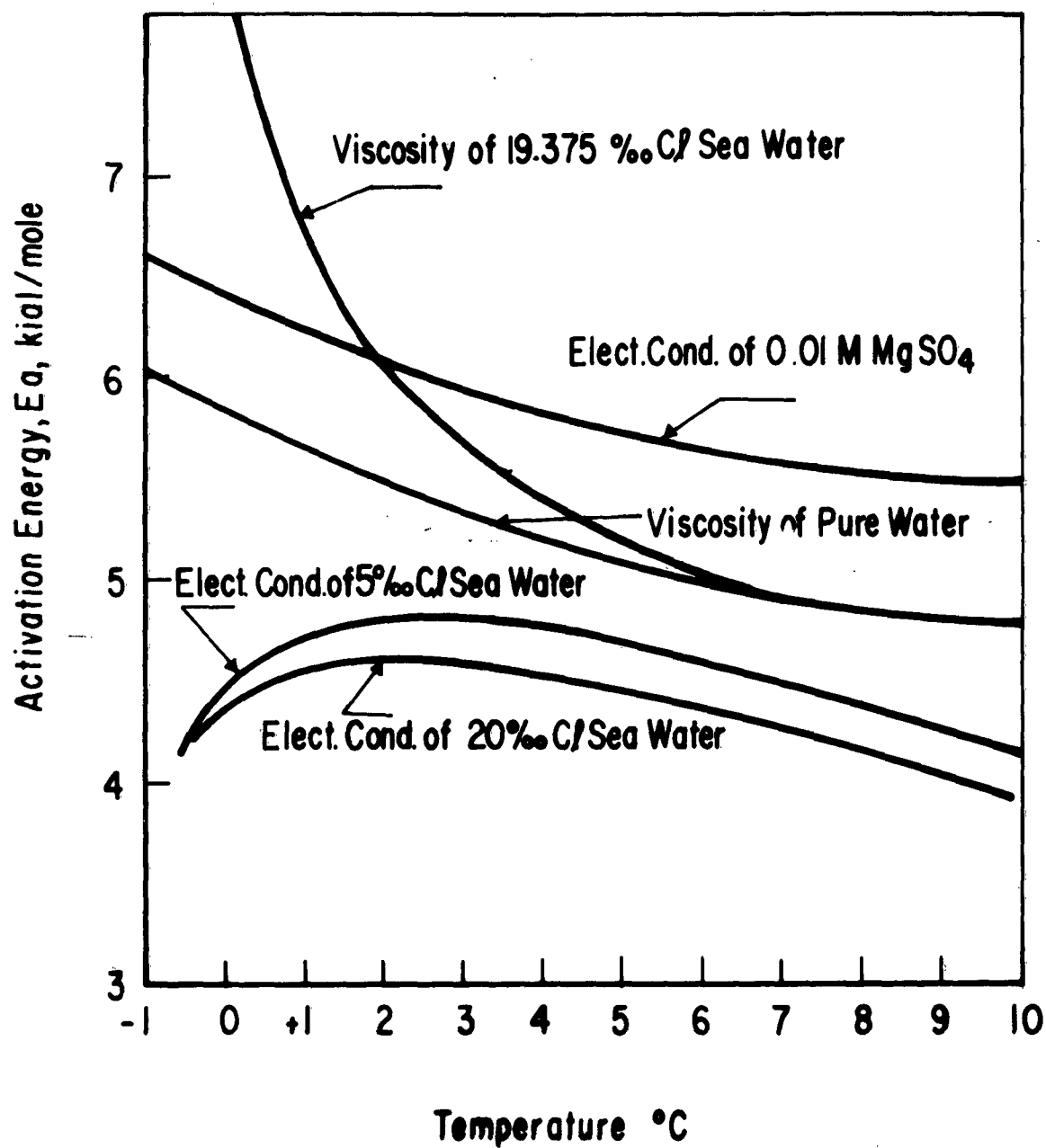
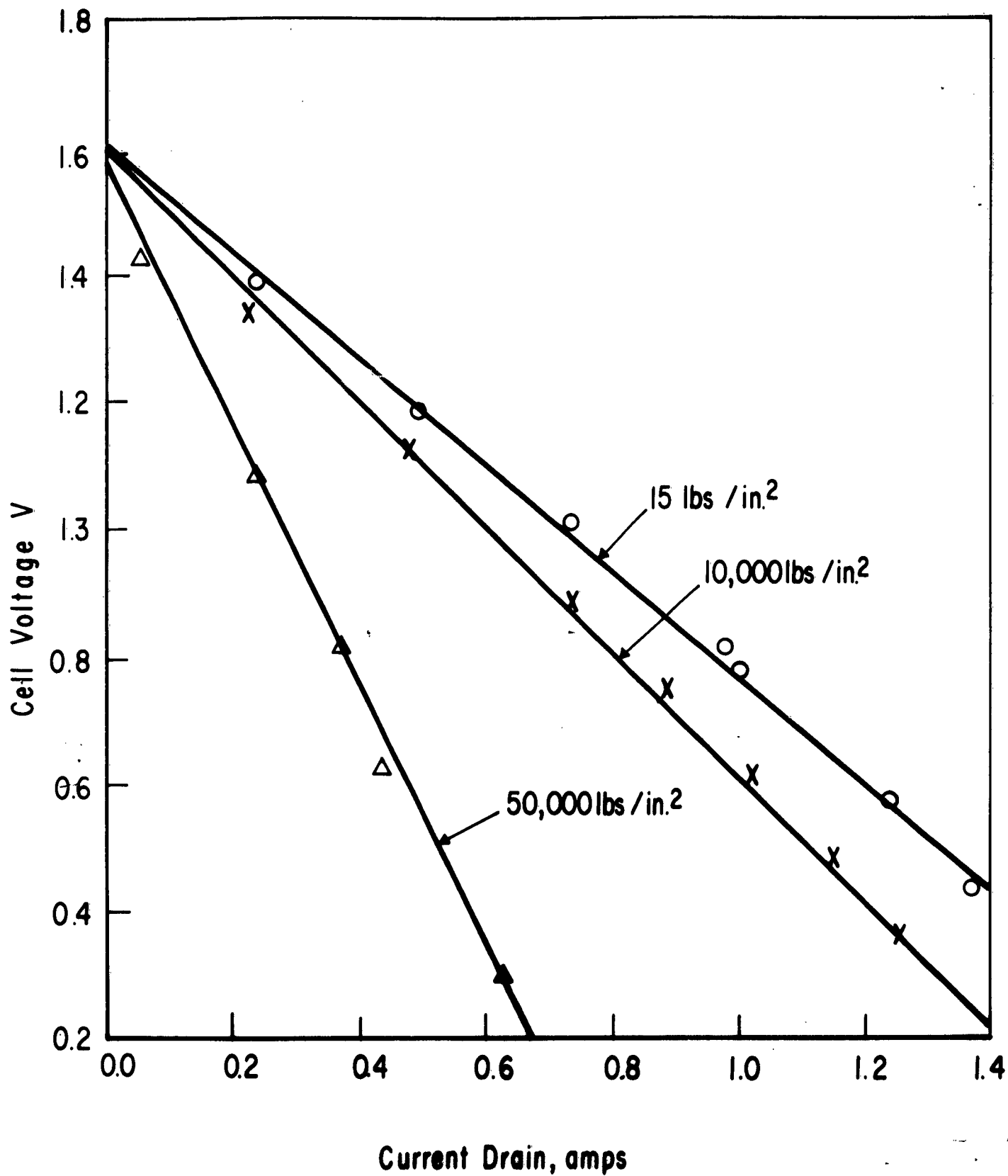


Figure 6

Discharge Characteristics of an Eveready
Size AA Dry Cell at 25°C Under Pressure



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		2b. GROUP	
3. REPORT TITLE Physical Chemistry of the Deep Ocean Environment			
4. DESCRIPTIVE NOTES (Type of report and inclusive dates) Technical Report			
5. AUTHOR(S) (Last name, first name, initial) Horne, R. A.			
6. REPORT DATE May 1, 1965		7a. TOTAL NO. OF PAGES 23	7b. NO. OF REFS 23
8a. CONTRACT OR GRANT NO. Nonr-4424(00)		9a. ORIGINATOR'S REPORT NUMBER(S) 11	
b. PROJECT NO. Task No. NR051-460		9b. OTHER REPORT NO(S) (Any other numbers that may be assigned this report)	
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